

Effect of inorganic salts on Se(IV) and Re(VII) diffusions in bentonite*

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Diffusion behaviors of Se(IV) and Re(VII) in bentonite were investigated by a through-diffusion method in nitrate, sulfate, carbonate and silicate solutions. SEM-EDS analysis showed that Se(IV) was reduced to red precipitate Se(0) by sulfite. Se(IV) was sorbed on bentonite with distribution coefficient K_d of $(2.6-5.3) \times 10^{-4}$ m³/kg in sulfite, nitrate and sulfate solutions, whereas it was hardly sorbed in carbonate and silicate solutions. The effective diffusion coefficients were $D_e = (0.81-7.0) \times 10^{-11}$ m²/s for Se(IV) and $D_e = (1.4-4.4) \times 10^{-11}$ m²/s for Re(VII). The D_e value of Se(IV) exhibited a dependence on the inorganic salts in the order of sulfite \approx nitrate \approx sulfate $>$ silicate $>$ carbonate, whereas the salts had insignificant effect on Re(VII) diffusion. The results suggest that the discrepancy in diffusion mechanism may lead to the different impact of the salts on the diffusion of Se(IV) and Re(VII) in GMZ bentonite.

Keywords: Diffusion, Selenite, Perrhenate, Competition effect, Bentonite

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I. INTRODUCTION

In safety assessment of HLW (high-level radioactive waste) repository, the long-lived radionuclides ⁷⁹Se (6.5×10^4 y) and ⁹⁹Tc (2.13×10^5 y), which exist predominately as anions in oxidizing aqueous environment, are identified to be among the largest contributors to the global radioactivity that can be released from high level radioactive waste repository to geosphere [1, 2], hence the importance of studying their sorption and diffusion behaviors. As a redox sensitive element, selenium exists four oxidation states: selenide, Se(II); elementary selenium, Se(0); selenite, Se(IV); and selenate, Se(VI). Under mild redox condition, anion Se(IV) is the predominately species in aqueous solution, which is among the main concerned species in safety assessment of repository.

Normally, anions have high mobility in most of geological materials with negative surface because of anion exclusion, but Se(IV) can be sorbed on many materials, such as clay minerals [3, 4], iron oxides [5, 6] and rocks [7, 8]. In bentonite, composed mainly of montmorillonite, Se(IV) can be sorbed on Ca-montmorillonite under acid and neutral conditions by ligand exchange with reactive surface aluminol groups, located at the particle edges below pH 7 [9]. According to the investigation of Shi *et al.* [10], the sorption of Se(IV) on Gao-miaozi (GMZ) bentonite in the pH range of 3–9 could be explained by a surface complexation reaction between HSeO₃⁻ and surface aluminol groups. Boulton *et al.* [11] investigated the sorption of Se(IV) on Kunigel-V1 bentonite and suggested that minor phases containing iron were responsible for sorption above pH 7. EXAFS study shows that the sorption

of Se(IV) on pure montmorillonite forms bidentate binuclear inner-sphere complexation. For Al hydroxide and aluminosilicate minerals, a mixture of outer-sphere and bidentate binuclear inner-sphere is observed [12].

Being in the same group in the periodic table, rhenium and technetium have similar reactivity. Technetium is air-sensitive, the insoluble Tc(IV) can be easily oxidized to high mobility Tc(VII) [13]. In order to avoid the complicated operation of radioactive experiments, rhenium is often used as an analog for ⁹⁹Tc. Our recent work showed that Re(VII) had similar effective diffusion coefficient (D_e) to Tc(VII) in bentonite. Therefore, it can also be used as the analog for ⁹⁹Tc in diffusion study [14].

In deep underground storage of HLW in China, GMZ bentonite is chosen as the candidate backfilling material [15]. Beishan Mountain is chosen as the potential HLW repository site, where the underground water contains a large amount of sulfate and carbonate [16]. Therefore, sulfate, carbonate and silicate species can be present in noticeable quantities in interstitial water. The competition effect of major co-existing anions has been extensively discussed by many authors [17–19]. Wellman *et al.* [20] found that the Re release in carbonate concrete monoliths was 1–3 order of magnitudes higher than that non-carbonated ones. Carbonate hindered the Re sorption by means of the negated reduction effect of iron on Re. In the case of Se(IV), the inorganic anions decrease the retention capacities on the sorption sites of clay/rock materials [7], iron oxides [21, 22] and manganese oxides [23, 24]. For examples, silicate and carbonate hinder the sorption of Se(IV) onto magnetite [25], and, silicic acid decreases its sorption capacity onto magnetite and hematite [21, 22]. In addition, silicate hinders the Se(IV) sorption on goethite significantly when the concentration ratio of silicate/Se(IV) is over 100, whereas the effect of sulfate is found to be insignificant [26]. High concentration of sulfate is found to reduce the sorption

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TABLE 1. The chemical components of GMZ bentonite [15]

Salts	Al ₂ O ₃	SiO ₂	P ₂ O ₅	CaO	K ₂ O	TiO ₂	FeO	TFe ₂ O ₃	MgO	Na ₂ O	MnO	Loss on ignition
Content (wt.%)	14.20	67.43	0.02	1.13	0.73	0.12	0.29	2.40	0.10	1.75	0.02	11.38

of Se(IV) on hematite and magnetite [7]. However, Goh *et al.* [17] found that the sulfate effect on Se(IV) sorption was negligible on iron-bearing soils in the tropical zones, even with an increased concentration of sulfate.

As far as we know, little information has been published concerning the effect of inorganic anions on the Se(IV) and Re(VII) diffusion in bentonite, and diffusion method has not been used to measure the corresponding diffusion parameters. In this work, a through-diffusion method was employed to measure the D_e and rock capacity factor α values of Se(IV) and Re(VII) in background solutions (sulfite, nitrate, sulfate, carbonate and silicate). Se(IV) and Re(VII) were chosen as the represented oxyanions for divalent and monovalent anions. The results can help to better understand the behaviors of anions in a radioactive waste repository.

II. MATERIALS AND METHODS

A. Materials

The GMZ bentonite (75.4 wt.% montmorillonite) was obtained from Beijing Research Institute of Uranium Geology as a gift without any pretreatment. It was compacted to cylinder of $\Phi 2.54 \text{ cm} \times 0.9 \text{ cm}$. The dry density was 1600 kg/m^3 . The chemical components presented by Ye *et al.* [15] are listed in Table 1.

All chemicals used in this study were analytical grade and all solutions were prepared using deionized water. A stock solution containing Se(IV), Re(VII) and five different inorganic anions were prepared in deionized water. The background solutions included sulfite, sulfate, nitrate, carbonate and silicate, which were prepared by dissolving analytical grade chemicals of Na_2SO_3 , Na_2SO_4 , NaNO_3 , Na_2CO_3 and Na_4SiO_4 in deionized water, respectively. The concentration of background anions was 0.05 mol/L . The solution was added with 0.003 mol/L NaN_3 to avoid bacterial growth. The diffusion cells were put in an incubator to maintain the temperature at $(40 \pm 2)^\circ\text{C}$. The initial concentration of 3000 mg/L Se(IV) and 650 mg/L Re(VII) were prepared by dissolving SeO_2 (from Sinopharm Reagent) and NH_4ReO_4 (Alfa Aesar, 99.997%). The concentration of Se(IV) and Re(VII) were determined by ICP-OES (PerkinElmer Optima 2100DV). The selenium powder was examined by a scanning electron microscope (Hitachi S-3400N) equipped with an energy dispersive X-ray spectroscopy (EDS).

B. Diffusion experiments

The diffusion set-up and experimental procedures were described by in Ref. [27]. After the bentonite sample

was saturated by 0.05 mol/L inorganic salt solutions for 5 weeks, Se(IV) and Re(VII) were introduced simultaneously in 200 mL source reservoir. The target reservoirs contained 10 mL solution with the 0.05 mol/L different inorganic salt solutions were regularly replaced after a given time interval. The pH and Eh (PB-100, Sartorius, Germany) were measured in the source and target reservoirs so as to get information of the species of Se(IV) and Re(VII).

C. Diffusion experiments

For the one-dimensional diffusion process, Fick's second law was applied. Best-fit parameter values for D_e and α were obtained using Eq. (1), which represents the diffusion through a planar sheet:

$$A_{\text{cum}} = S \cdot L \cdot C_0 \cdot \left(\frac{D_e t}{L^2} - \frac{\alpha}{6} - \frac{2\alpha}{\pi^2} \sum_{n=1}^{\infty} \frac{(-1)^n}{n^2} \exp \left\{ -\frac{D_e n^2 \pi^2 t}{L^2 \alpha} \right\} \right), \quad (1)$$

where, L is the sample thickness (in m); C_0 (in $\mu\text{g/m}^3$) is the initial concentration of the diffusing species; A_{cum} (in μg) is the total amount of diffused species through a boundary at $x = L$ and at diffusion time t ; and S is the cross section area (in m^2) of the clay. The flux $J(L, t)$ at the low-concentration boundary ($x = L$) is calculated by Eq. (2):

$$J(L, t) = \frac{1}{S} \cdot \frac{\partial A}{\partial t}. \quad (2)$$

The experimental and theoretical data processing in through-diffusion methods have been described previously [27]. Simulation of the experiment was accomplished by computer code compiled by Mathematica 6.0 [28].

III. RESULTS AND DISCUSSION

A. Diffusion behavior of Se(IV)

Figure 1 shows flux $J(L, t)$ and accumulated mass A_{cum} as function of diffusion time for Se(IV) in presence of sulfite. The measured A_{cum} (solid dots) agree well with the solid line calculated by Eq. (1), and the $J(L, t)$ measured (empty dots) agree well with the dotted line calculated by Eq. (2). Anion diffusion follows Fick's second law, which undergoes the transient- and steady-state phases. In transient-state phase, the flux increases dramatically and the A_{cum} increases slowly. While, the flux remains unchanged and A_{cum} increases linearly in steady-phase. It took ca. 8 days to reach steady-state phase for Se(IV) in presence of sulfite.

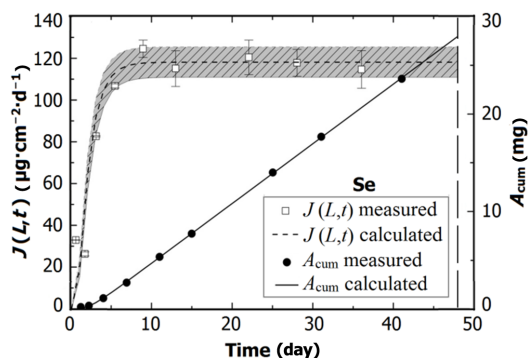
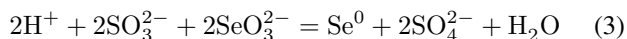


Fig. 1. A_{cum} and flux of Se(IV) diffusion in GMZ bentonite in sulfite solution.

Sulfite might exist under redox condition in deep repository. Since selenium lies in the same group with sulfur in the periodic table, SeO_3^{2-} may be reduced by SO_3^{2-} . However, to our knowledge, the redox ability of SO_3^{2-} to SeO_3^{2-} has not been reported under ambient condition. Similar mass of Se(IV) was added in the source reservoirs, the initial concentration of sulfite solution was lower than that of the others. A red dark precipitate was found on the surface of glass source reservoir. It was measured by TEM-EDS. Figure 2(a) shows the SEM image of flake-like precipitate. From EDS analysis (Fig. 2(b)), the precipitate consists of Se (93.5%), C (2.3%), O (3.0%), S (0.4%) and Na (0.8%). The standard reduction potential of SO_3^{2-} is lower than that of SeO_3^{2-} , i.e., $E^\ominus(\text{SeO}_3^{2-}/\text{SeO}_4^{2-}) = -0.056 \text{ V}$, and $E^\ominus(\text{SeO}_3^{2-}/\text{Se}) = 0.74 \text{ V}$. It can be explained that Se(IV) is reduced to the precipitate Se(0) by sulfite.



Nitrate often occurs in large amounts in interstitial water in repository. Kim *et al.* [25] found that there was insignificant difference in the sorption of Se(IV) on magnetite among the solution with nitrate and perchlorate, without the electrolytes. Montavon *et al.* [4] also found that nitrate, sulfate and chloride had neglected effect on the sorption of Se(IV) on bentonite. Figure 3 shows the breakthrough curves for Se(IV) in sulfite, nitrate, sulfate, carbonate and silicate solutions, respectively. The modeling curves fit well with experimental data for all the inorganic salts. The A_{cum} exhibited a dependence on the inorganic salts in the order of sulfite \approx nitrate \approx sulfate $>$ silicate \approx carbonate. It indicates that silicate and carbonate hinder the diffusion. The breakthrough curves were similar in nitrate and sulfate solutions. It could be deduced that sulfate had insignificant effect on Se(IV) diffusion in GMZ bentonite. Similar results can be found elsewhere [4, 17, 26].

B. Diffusion behavior of Re(VII)

Figure 4 shows flux $J(L, t)$ and accumulated mass A_{cum} as a function of diffusion time for Re(VII) in presence of sulfite.

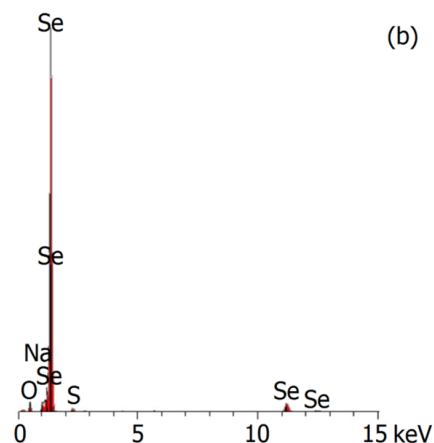
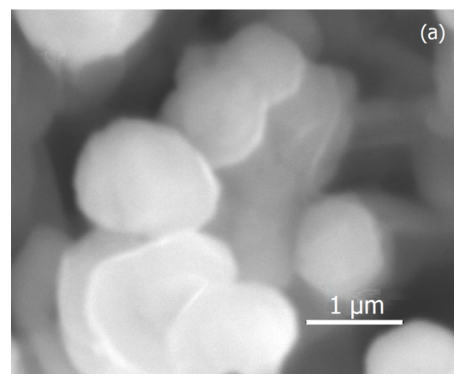


Fig. 2. (Color online) SEM image (a) and EDS spectrum (b) of Se(0) from SeO_3^{2-} reduction by sulfite.

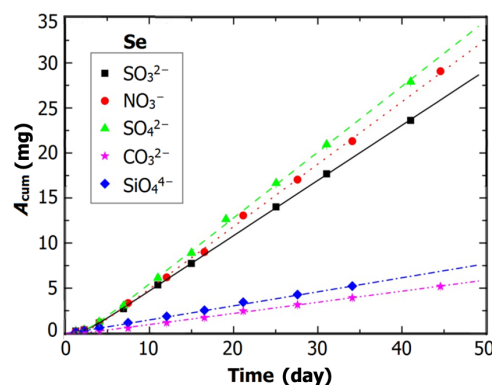


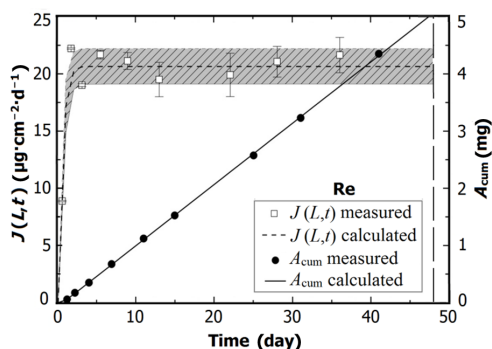
Fig. 3. (Color online) A_{cum} of Se(IV) diffusion in GMZ bentonite as a function of time in inorganic salt solutions. The lines are modeling curves.

It took ca. 2 days to reach steady-state phase, indicating that the diffusion of Re(VII) is faster than that of Se(IV). The initial concentration of Re(VII) in sulfite solution was the same as in the other four solutions, demonstrating that Re(VII) cannot be reduced by sulfite under ambient condition.

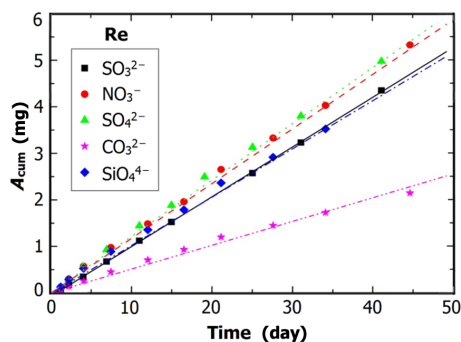
Figure 5 shows the breakthrough curves for Re(VII) in sulfite, nitrate, sulfate, carbonate and silicate solutions, respectively. The breakthrough curves were almost identical to those of Se(IV) except the silicate solution. It implies that

TABLE 2. Anionic species effect on diffusion parameters for Se(IV) and Re(VII) in GMZ bentonite

pH	Inorganic salts	C_0 (mg/L)		$D_e (\times 10^{-11} \text{ m}^2/\text{s})$		α		$K_d (\times 10^{-4} \text{ m}^3/\text{kg})$	
		Se(IV)	Re(VII)	Se(IV)	Re(VII)	Se(IV)	Re(VII)	Se(IV)	Re(VII)
2.5	SO_3^{2-}	2400 ± 100	650 ± 20	7.0 ± 0.9	4.4 ± 0.4	1.28 ± 0.03	0.14 ± 0.04	5.3 ± 0.3	-1.9 ± 0.5
2.3	NO_3^-	3000 ± 100	660 ± 20	4.5 ± 0.5	3.5 ± 0.2	0.88 ± 0.05	0.012 ± 0.004	2.8 ± 0.2	-2.7 ± 0.9
2.5	SO_4^{2-}	2800 ± 100	650 ± 20	5.1 ± 0.6	3.7 ± 0.2	0.86 ± 0.05	0.015 ± 0.004	2.6 ± 0.2	-2.7 ± 0.7
8.5	CO_3^{2-}	3000 ± 100	670 ± 20	0.81 ± 0.09	1.4 ± 0.2	0.13 ± 0.02	0.019 ± 0.003	-1.9 ± 0.3	-2.6 ± 0.4
10	SiO_4^{4-}	3000 ± 100	680 ± 20	1.02 ± 0.03	2.7 ± 0.2	0.039 ± 0.003	0.042 ± 0.005	-2.5 ± 0.2	-2.5 ± 0.3

Fig. 4. A_{cum} and flux of Re(VII) diffusion in GMZ bentonite in sulfite solution.

carbonate hinders the Re(VII) diffusion. The pH of silicate solution was 10.0, and those of sulfite, nitrate and sulfate solutions were around 2.5. Their breakthrough curves were almost identical, indicating that the impact of pH on the mobility of Re(VII) can be neglected.

Fig. 5. (Color online) A_{cum} of Re(VII) diffusion in GMZ bentonite as a function of time in inorganic salt solutions. The lines are modeling curves.

C. Diffusion parameters of Se(IV) and Re(VII)

Table 2 lists the effect of the inorganic salt solutions on the diffusion parameters of Se(IV) and Re(VII). The D_e value of Se(IV) exhibited a dependence on the inorganic salt solutions in the order of sulfite \approx nitrate \approx sulfate $>$ silicate $>$ carbonate. For sulfite, nitrate and sulfate solutions, the α values were higher than the total porosity ε ($\varepsilon = 0.44 \pm 0.02$), which

was measured by the HTO diffusion in GMZ bentonite [29]. The distribution coefficient K_d and its uncertainty are calculated by Eqs. (4) and (5), respectively:

$$K_d = (\alpha - \varepsilon)/\rho, \quad (4)$$

$$uK_d = K_d \{ [r.u(\alpha)]^2 + [r.u(\varepsilon)]^2 + [r.u(\rho)]^2 \}^{1/2}, \quad (5)$$

where ρ is the dry density (in kg/m^3). The K_d value was around $10^{-4} \text{ m}^3/\text{kg}$, being one order of magnitude lower than that obtained by sorption experiments [4]. The reason could be the discrepancy of the chemical compounds in MX-80 and GMZ bentonites, or the difference between the batch methods conducted the powder bentonite sample measured by Montavon *et al.* [4] and the through-diffusion method conducted the compacted ones in this work. In the case of carbonate and silicate solutions, α was less than ε , indicating that Se(IV) cannot be sorbed on bentonite. Similar results can be found elsewhere [21, 22, 25]. In the case of Re(VII), the difference in D_e value was within two times. It implies that the impact of inorganic salts on Re(VII) diffusion can be neglected except carbonate. Besides, K_d values were negative, indicating that Re(VII) cannot be sorbed on the negatively charged surface of bentonite because of anion exclusion.

IV. CONCLUSION

The diffusion behaviors of Se(IV) and Re(VII) in Gao-miaozi (GMZ) bentonite were investigated by the through-diffusion method in sulfite, nitrate, sulfate, carbonate and silicate solutions. In the case of sulfite solution, a red dark precipitate was found on the surface of glass source reservoir, with decreased initial concentration of Se(IV). The precipitate was identified to be Se(0) by SEM-EDS, indicating that Se(IV) was reduced by sulfite under ambient condition. Both Se(IV) and Re(VII) diffusions were unaltered in nitrate and sulfate solutions. Both the D_e and α values of Se(IV) decreased in carbonate and silicate solutions, indicating that they hinder Se(IV) diffusion. The inorganic salts except carbonate had insignificant effect on the Re(VII) diffusion.

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